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THE JOHNS HOPKINS UNIVERSITY
APPLIED PHYSICS LABORATORY
8621 Georgia Avenue,   Silver Spring, Maryland

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METHANE-OXYGEN FLAME STRUCTURE

CONSERVATION OF MATTER AND ENERGY IN THE ONE-TENTH ATMOSPHERE FLAME

by

A. A. Westenberg and R. M. Fristrom

January 1960

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ABSTRACT

The experimental data on temperature, composition, and aerodynamic profiles through a flat, premixed CH_4-O_2 flame obtained by techniques described in the report CM-968, Characteristic Profiles in a Low-Pressure, Laminar, Lean, Premixed Methane-Oxygen Flame, of this series are analyzed in terms of matter and energy conservation. The composition profiles for the seven stable species found in the flame are corrected for molecular diffusion, using diffusion coefficients measured in this Laboratory, and the resulting flux distributions are shown to give a satisfactory balance of carbon and hydrogen through the flame. The fluxes of enthalpy by means of convection, diffusion, and conduction are given separately and summed to show the degree to which energy conservation is fulfilled by the data. The various approximations used in the treatment are critically discussed, and it is shown that the general reliability of the results has been considerably improved over previous laminar flame structure analyses.

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ATMOSPHERE FLAME

I. INTRODUCTION

The development of experimental techniques for the measurement of temperature, composition, and aerodynamic profiles through laminar flame zones has been reported in earlier publications of a series from the Applied Physics Laboratory (Refs. 1, 2 and 3). This work culminated in a set of data obtained in a stoichiometric, premixed, propane-air flame of conical geometry at 1/4 atmosphere pressure. The fourth paper of the Combustion and Flame series (Ref. 4) reported the analysis of these data in terms of material transport.

The experience gained up to that time pointed the way to various refinements and changes in the experimental techniques, and also to the desirability of studying a flame system which was chemically simpler insofar as the number of stable species present is concerned. The changes in technique were described in detail in Ref. 5. In brief summary, these included the following:

1. A flat-flame stabilized on a screen burner was used instead of the conical flame used in the previous work. This brought about some simplification in the geometrical handling of the data, and in the physical manipulation of the apparatus.

References may be found on pages 23, 24, and 25.

2. The operating pressure was lower (0.1 atmosphere instead of 0.25 atmosphere) which gave a thicker flame and hence eased the requirements on spatial resolution.

3. Gas sampling and mass spectrometer analyses were done in a continuous flow system instead of batchwise. This afforded much better precision, and circumvented wall adsorption difficulties. In particular, this system permitted good analyses for water to be realized.

4. Temperature profiles were obtained with a fine, silica-coated Pt-Pt, 10 per cent Rh thermocouple rather than by particle-track photography (which was used only as a supplementary technique). Considerably better precision was thus attained.

5. Improvements in controlling and monitoring the flow of gases to the burner were introduced which led to better flame stability. A monitoring thermocouple was mounted just downstream of the screen, which allowed the flame position to be monitored at all times.

The chemically simpler flame system finally decided on was a mixture of methane and oxygen. It was thought that this represented a reasonable compromise between chemical simplicity, ease of handling, and some degree of general interest as a common combustible mixture. As shown in Ref. 5, this flame contained five major stable species (methane, oxygen, carbon monoxide, carbon dioxide, and water) and two minor ones (hydrogen and formaldehyde) for which analyses were made, which is considerably simpler than the twelve components found in the propane-air flame. No other species

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(other than impurities) at a concentration greater than
0.01 per cent were found.

One other important improvement incorporated in this
new work should be mentioned. This is that experimentally
determined molecular diffusion coefficients were used in the
data analysis to be described. In view of the pronounced
(in some cases the predominant) effect of diffusion on the
flux profiles of the various flame constituents, this is a
decided gain in the reliability of this general approach to
laminar flame studies, particularly in the derivation of
chemical kinetic information from the data. In the previous
work on propane-air flames, it was necessary to use values
of the molecular diffusion coefficients calculated from
kinetic theory and empirical low temperature viscosity
parameters, so that the reliability of the diffusion coef-
ficients at flame temperatures was quite uncertain. Since
then a new method of measuring diffusion coefficients up
to moderately high temperatures ($\sim 1200^\circ$K) has been devel-
oped at the ...lied Physics Laboratory (Refs. 6, 7 and 8)
and used specifically to measure the pertinent coeffi-
cients for the methane-oxygen flame (Ref. 9).

The present paper describes the analysis of the data
on the methane-oxygen flame from the viewpoint of the trans-
fer of mass and energy. It is believed that the various
changes and refinements noted above have contributed to
making the analysis of this flame considerably more reliable
than was possible before.

II.   PREPARATION OF THE DATA

As described in detail in Ref. 5, the first paper of this series, the flame used in this study was flat, and held at a pressure of 7.60 cm Hg. The initial mixture composition (mole fractions) aside from traces of impurities totaling less than 1 per cent (argon, nitrogen, carbon dioxide, and water) was as follows:

$$x_{CH_4} = 0.0785 ; \qquad x_{O_2} = 0.9143.$$

The data finally used in the subsequent analysis consisted of a set of concentration profiles for the stable species present, a thermocouple temperature profile, a streamtube area ratio profile, and a mass average gas velocity profile. The latter was obtained from a measurement of the final (hot boundary) gas velocity v_f by means of particle-track photography and the temperature profile, making use of the continuity relation

$$\rho_f v_f a_f = \rho v a , \qquad (1)$$

where ρ is density and a is the cross-sectional area of a streamtube, and the equation of state

$$\rho = P\bar{M}/RT , \qquad (2)$$

where $\bar{M}$ is the mean molecular weight and the other symbols are conventional. Since the thermocouple temperature profile was much smoother and more reproducible than the particle-track velocity profile, it was felt that this procedure of deriving the velocity from the temperature by fitting at the measured point where the particle-track data

were most reliable was preferable. Note that this is the
reverse procedure to that used previously (Ref. 4) where
the temperature profile was derived from particle-track
velocity measurements.

The problem then arises as to how best to super-
impose, spatially, the temperature profile (as discussed
above, the gas velocity and area ratio profiles were
coupled to the temperature, and thus not independently
variable) and the composition profiles determined by in-
dependent means. It has been pointed out (Ref. 1) that
when temperature and composition are determined with
different devices (thermocouple and sampling probe) it
leads to an uncertainty in referring both to a common
spatial origin even though the positions of both are
measured relative to a fixed point on the burner (the moni-
toring thermocouple). The pneumatic (sonic flow) probe
temperature measurements were taken with the same probe as
used in sampling, consequently it was assumed that a tem-
perature read in this way would coincide automatically with
the appropriate composition point. Thus the method used to
determine the absolute origin was to superimpose the ther-
mocouple and pneumatic probe temperature "knees", that is,
the point of intersection of straight lines extrapolated
from the hot gas region and the region of rapid rise in the
temperature profiles. The composition profile was then fixed
relative to the thermocouple temperature profile. Additional
discussion of this problem is included in the next section.
The distance coordinate z was taken to be zero at the moni-
toring thermocouple just downstream of the screen.

III. ANALYSIS OF THE DATA

Conservation of Matter

With the primary data on composition, temperature, and mass average gas velocity available, the first step in the analysis was to take account of diffusion effects. The quantity G_i is the fraction of the total mass flow at any point in the flame which is due to chemical species i, and is given by

$$G_i = f_i(v+V_i)/v \, , \qquad\qquad (3)$$

where f_i is the concentration expressed as mass fraction at any point and V_i is the diffusion velocity. It is a net flux variable which includes a positive contribution from diffusion if the concentration of i decreases downstream, or a negative contribution in the opposite case. As in the previous work (Ref. 4), the diffusion velocity V_i was calculated by assuming first, that each species i could be regarded as a trace component in the mixture and, second, (since oxygen is present in large excess everywhere) each species i could be treated as being in a binary mixture with oxygen (see Appendix A). Under these conditions the approximate relation

$$V_i = - (D_i/x_i)(dx_i/dz) \qquad\qquad (4)$$

may be used, where D_i is the binary diffusion coefficient with oxygen. (This also neglects the effect of thermal diffusion, a point which is discussed in Appendix B.) Equation (4) was used to compute the V_i for all the flame species (except oxygen, of course) at each point throughout

the flame. The diffusion velocity for oxygen was then
obtained from the required normalization condition

$$\sum_i N_i \, M_i \, V_i = 0 \qquad\qquad (5)$$

(N_i is the molar density of species i, and M_i its molec-
ular weight) which must be fulfilled in view of the defi-
nition of diffusion velocity (relative to the mass aver-
age gas velocity v).

The concentration gradients required in Eq. (4) for
each species were obtained by numerical differentiation of
the smooth curves drawn through the experimental data, such
as are shown as the solid curves in Figs. 1 through 7. The
numerical process was supplemented by graphical differen-
tiation at those portions of the curve where the slope
changed rapidly. The internal consistency of the differen-
tiations was checked. using the requirement that
$\sum_i (dx_i/dz) = 0$, since $\sum_i x_i = 1$.

The diffusion coefficients D_i used in Eq.(4) were
measured as a function of temperature in this Laboratory
(Ref.9). The point-source technique used permitted meas-
urement of the gas pairs CO_2-O_2 and H_2O-O_2 up to about
$1100^O K$, while CO-O_2, H_2-O_2, and CH_4-O_2 were limited to
slightly lower temperatures because of self-ignition. In
all cases, however, the temperature range covered was
enough to permit fitting the data to the appropriate kinetic
theory formulas (Ref. 9), and reliable extrapolation to the
highest temperatures needed in this flame ($2000^O K$). Dif-
fusion coefficients for the CH_2O-O_2 pair were not measured.
For this minor component, the necessary values were ob-
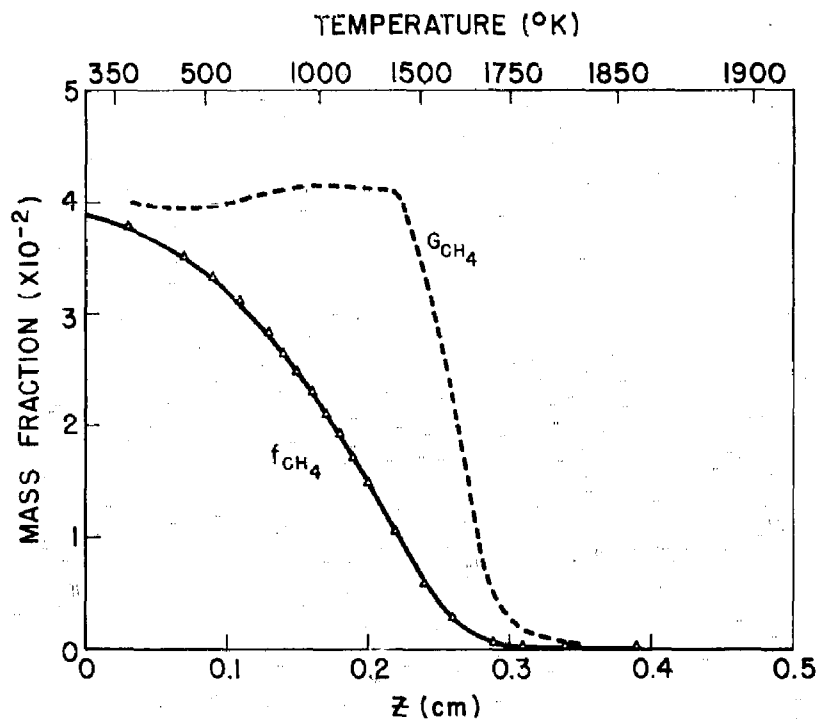tained from kinetic theory (Ref. 10) using the Lennard-

TEMPERATURE (°K)

Fig. 1 CONCENTRATION (f) AND FLUX FRACTION (G)
PROFILES OF METHANE THROUGH FLAME ZONE
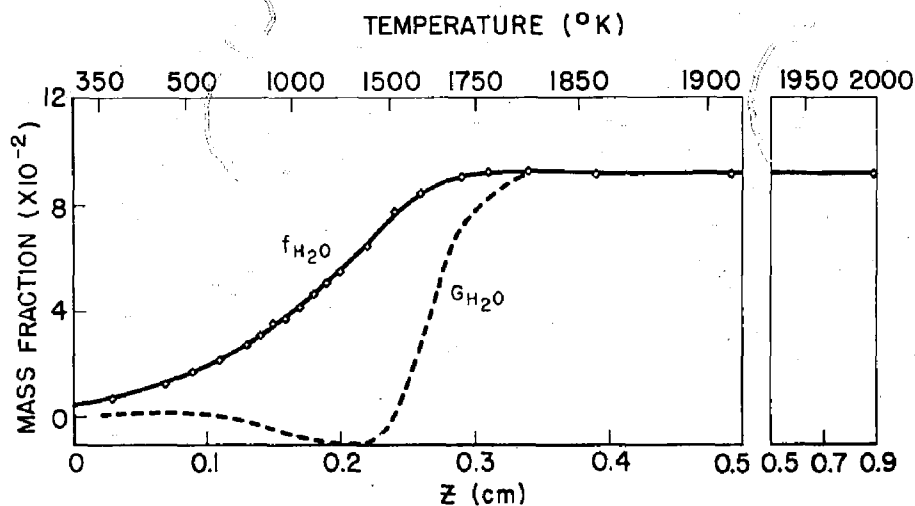
TEMPERATURE (°K)

Fig. 2 CONCENTRATION (f) AND FLUX FRACTION (G)
PROFILES OF WATER THROUGH FLAME ZONE

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Fig. 3   CONCENTRATION (f) AND FLUX FRACTION (G)
PROFILES OF CARBON DIOXIDE THROUGH FLAME ZONE

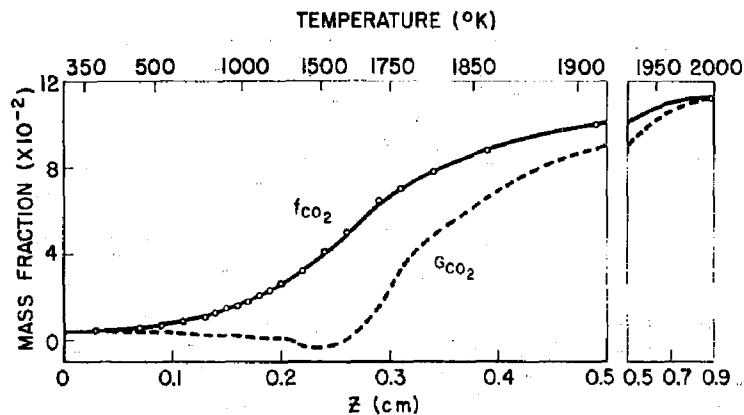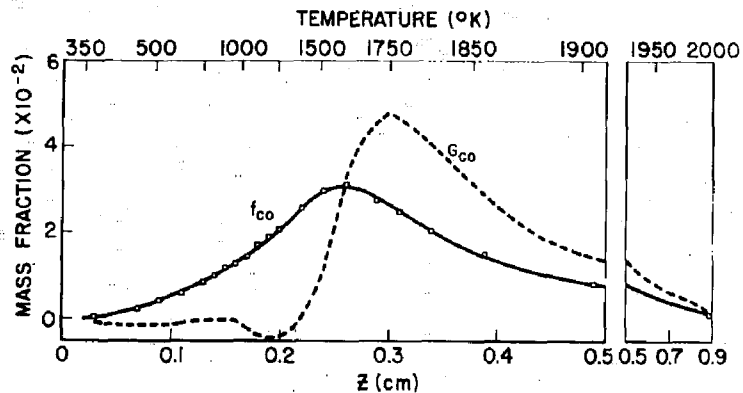Fig. 4   CONCENTRATION (f) AND FLUX FRACTION (G)
PROFILES OF CARBON MONOXIDE THROUGH FLAME ZONE

Fig. 5   CONCENTRATION (f) AND FLUX FRACTION (G)
PROFILES OF HYDROGEN THROUGH FLAME ZONE

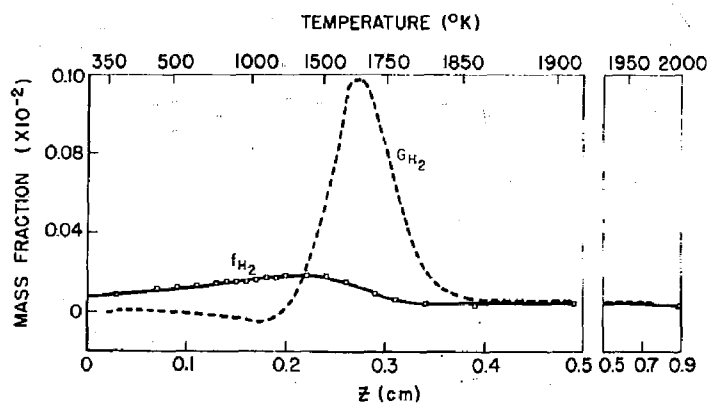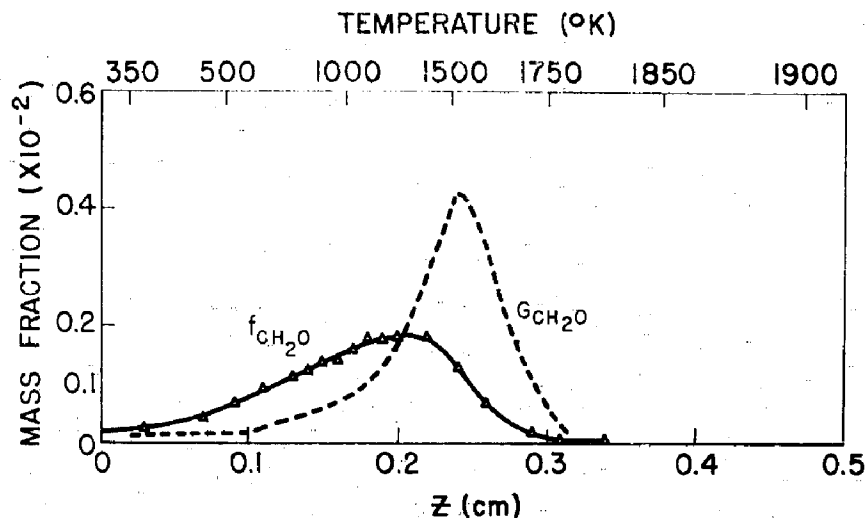TEMPERATURE (°K)

Fig. 6   CONCENTRATION (f) AND FLUX FRACTION (G)
PROFILES OF FORMALDEHYDE THROUGH FLAME ZONE
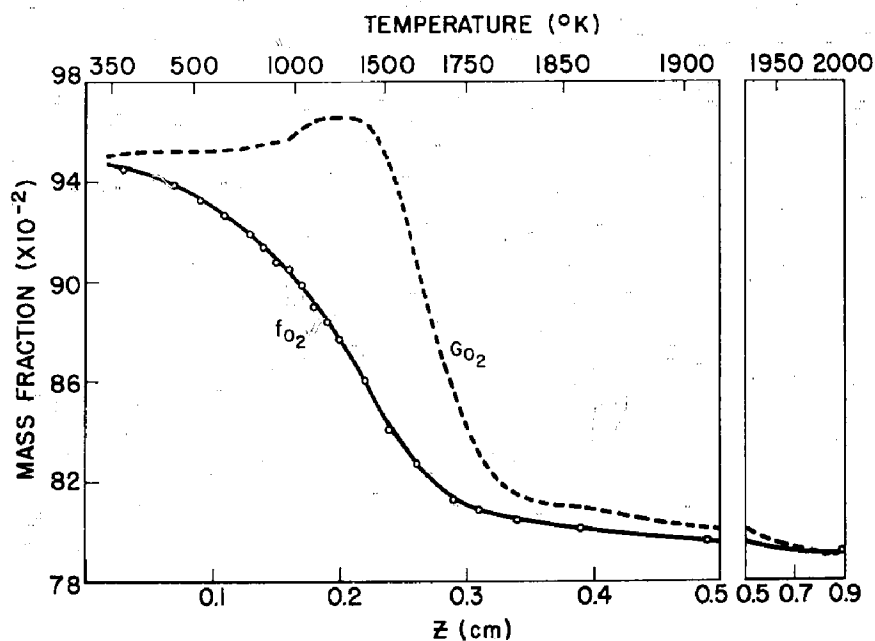
TEMPERATURE (°K)

Fig. 7   CONCENTRATION (f) AND FLUX FRACTION (G)
PROFILES OF OXYGEN THROUGH FLAME ZONE

Jones (6-12) potential and viscosity parameters. Since the latter are unavailable for formaldehyde, the values for methanol were used, this being a closely related molecule of similar size and mass. Table I lists the values of D_i versus temperature. The underlined values were extrapolated using the Lennard-Jones (6-12) potential with parameters fitted to the lower temperature data.

Table I

Binary Diffusion Coefficients as Function of Temperature. Underlined values are extrapolated; CH_2O-O_2 values computed from theory; pressure = 1 atmosphere.

	$D(cm^2/sec)$					
$T(^oK)$	CH_4-O_2	CO_2-O_2	H_2O-O_2	H_2-O_2	$CO-O_2$	CH_2O-O_2
300	0.226	0.161	0.288	0.821	0.224	0.180
500	0.581	0.419	0.692	2.09	0.542	0.422
700	1.05	0.767	1.22	3.76	0.956	0.739
900	1.63	1.19	1.85	5.79	1.45	1.12
1100	2.29	1.68	2.58	8.14	2.03	1.57
1300	3.03	2.23	3.41	10.8	2.68	2.07
1500	3.86	2.84	4.32	13.7	3.40	2.63
1700	4.75	3.50	5.32	16.9	4.18	3.24
1900	5.72	4.21	6.40	20.3	5.03	3.91
2100	6.76	4.98	7.55	24.0	5.94	4.61

The mass flux fractions G_i, having all the diffusion velocityes V_i at each 0.02 cm interval throughout the flame, were computed from Eq. (3). These are shown as the broken curves in Figs. 1 through 7. The great importance of diffusion is obvious in all cases. It will be noted that some of the G_i curves (water, carbon dioxide, carbon monoxide,

and hydrogen) have regions of negative values, which would
indicate a net upstream flux of these species. While there
does not appear to be any a priori reason why this is not
possible, the negative gradient dG_i/dz which exists up-
stream of these minima also implies that these species are
undergoing a net disappearance due to chemical reaction.
Since it is difficult to see how this could happen, it
seems likely that the negative G_i regions are not real and
should be regarded as errors in the over-all treatment. By
the same token, the positive maximum in the G_{O_2} probably
is not real since it is a direct consequence of the negative
G values for the other species by the normalization proce-
dure used to obtain the oxygen diffusion velocity.

As in the previous propane-air flame (Ref. 4) it is
important to examine the data in terms of conservation of
atomic species. If n_i is the number of atoms of a particular
element in a molecule of species i, it was shown that the
relation

$$\sum_i n_i G_i/M_i = \text{constant} \tag{6}$$

must hold throughout the flame, since atoms are neither
created nor destroyed. In the methane-oxygen flame, carbon
and hydrogen are the two elements whose conservation is to
be examined, the oxygen being present in such excess in the
form of O_2 that its constancy is practically assured. Re-
sults of applying Eq. (6) for carbon and hydrogen to the
G_i data given in Figs. 1 through 6 are shown in Fig. 8.
The data are given as per cent deviation from the initial
value of the summation, that is, the cold gas composition
upstream of the screen. (Because of diffusion, the probe
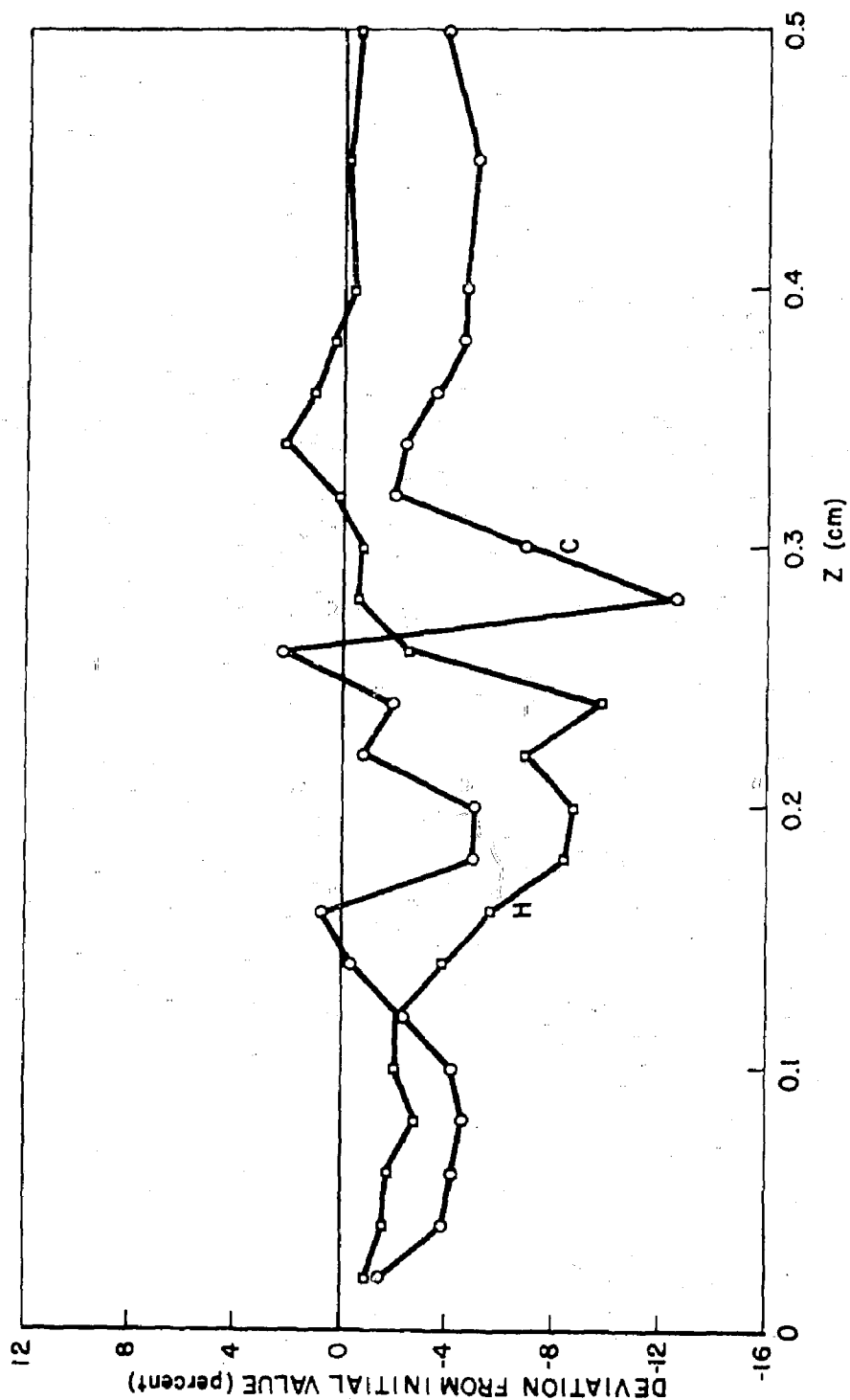composition just downstream of the screen at z = 0.03 cm

Fig. 8   CONSERVATION OF CARBON AND HYDROGEN (ATOM BALANCES) THROUGH FLAME ZONE

is not quite equal to the true cold gas composition.) The
balances are considered quite good, and are definitely better
than those in the earlier propane-air work. The worst de-
viation is -12 per cent for carbon and -9 per cent for hy-
drogen, while in most of the flame the balances are con-
siderably better (average deviations through the flame are
about -3 per cent). While the atom balance is not a very
sensitive test of the sampling data and the diffusion
corrections, the fact that it is reasonably good certainly
indicates there are no large errors in the data as a whole.

Conservation of Energy

It is of interest next to examine the data in terms
of energy considerations. Since the flame is an essentially
constant pressure system, the convenient energy variable to
use is the enthalpy. The energy continuity equation appro-
priate to a quasi-one-dimensional flow with area change such
as is found in the flat flame must first be derived.

The basic energy equation for a steady-state system
with no viscosity, radiation, or external forces may be
written

$$\nabla \cdot (\rho \, \vec{v} \, \hat{H} + \vec{q}) = 0 , \qquad (7)$$

where $\vec{v}$ is the mass average velocity vector, $\hat{H}$ is the
specific enthalpy of the mixture, and $\vec{q}$ is the heat flux
vector given by (page 717 of Ref. 10)

$$\vec{q} = -\lambda \nabla T + \sum_i N_i \, H_i \, \vec{V}_i . \qquad (8)$$

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if the inverse thermal diffusion (Dufour) effect is neg-
lected. λ is the mixture thermal conductivity, $\vec{V}_i$ is the
diffusion velocity vector, and H_i the absolute molar en-
thalpy of species i. The enthalpy per unit mass $\hat{H}$ is re-
lated to the H_i by

$$\hat{H} = \sum_i x_i H_i \Big/ \sum_i x_i M_i . \tag{9}$$

Making use of the theorem (page 815 of Ref. 10) which
states that, if $\vec{F}$ is a vector which is tangent to the sur-
face $f(x,y) = z$, then

$$\int (\nabla \cdot \vec{F}) \, dxdy = \frac{d}{dz} \int F_z \, dxdy ,$$

Eq. (7) may be converted to

$$\int (\rho \hat{H} v + q) \, dxdy = \text{constant}, \tag{10}$$

where v and q are the z-components (that is, normal to the
flame front) of their respective vectors. This may be
written as

$$\rho \, v \, a \, \hat{H} + q \, a = \text{constant}, \tag{11}$$

the quantities ρ, v, $\hat{H}$, and q now representing suitably
averaged values across the streamtube area a. Combining
the z-component of Eq. (8) with Eq. (11), and using the
quasi-one-dimensional over-all continuity relation in the
form

$$\rho_o \, v_o = \rho \, v \, A , \tag{12}$$

where $A = a/a_o$ and the subscript "$_o$" refers to the cold
boundary (that is, the screen), Eq. (11) becomes finally

$$\rho_o \, v_o \, \hat{H} - A \, \lambda (dT/dz) + A \sum_i N_i \, H_i \, V_i = \rho_o \, v_o \, \hat{H}_o , \tag{13}$$

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assuming that gradients in temperature and composition are essentially zero at the screen.

This equation represents the enthalpy conservation through the flame zone in terms of the flux of enthalpy per unit of inlet area at the screen. The first term on the right side is the flux due to convection, the second is that due to conduction, and the third that due to diffusion. These terms were evaluated for the experimental methane-oxygen data. The specific enthalpy required was obtained from Eq. (9) using tabulated molar enthalpies from standard sources (Refs. 11 and 12). The enthalpies for formaldehyde were obtained from the heat capacity data of Stevenson and Beach (Ref. 13). The thermal conductivity was taken to be that of pure oxygen, rather than attempting the exceedingly laborious computations for mixtures (for which all the necessary data are not available anyway). Because of the preponderance of oxygen, this approximation was adequate (see Appendix C). The thermal conductivity of oxygen has not been measured up to the temperature required here, so it was calculated by the following semi-empirical procedure: The translational (or monatomic) thermal conductivity λ_{mon} was obtained from the NBS tabulation (Ref. 11) of viscosity (η) by means of the rigorous theoretical (first approximation) relation

$$\lambda_{mon} = (15/4)(R/M)(\eta) .$$

The improved Eucken-type correction of Hirschfelder (Ref. 14) was then applied, that is

$$\lambda/\lambda_{mon} = 0.469 + 0.354 (C_p/R - 1) ,$$

to account for the effect of internal degrees of freedom. The temperature gradient was obtained by numerical

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differentiation of the temperature profile, and the other
quantities in Eq. (13) were already available.

The various enthalpy fluxes are shown in Fig. 9.
The conduction term is, of course, negative, since it
always represents a flux of heat (positive enthalpy) up-
stream. The diffusion term would be expected to be posi-
tive relative to the inlet flux as it is, since the react-
ants of high enthalpy diffuse downstream and the products
of low enthalpy diffuse upstream, both effects constituting
a net positive enthalpy flux. (It seems unlikely that the
effects of intermediates, which diffuse in both directions
depending on which side of their maximum in concentration
one considers, would alter this statement. They have en-
thalpies lower than the reactants and higher than the final
products.) Whether the convection term, that is, the spe-
cific enthalpy profile, constitutes a positive or negative
flux depends upon the relative magnitudes of the conduction
and diffusion terms, as has been noted by Lewis and von Elbe
(Ref. 15). These terms depend upon their respective trans-
port coefficients. In the special case (the theoreticians'
favorite) that all the binary diffusion coefficients are
equal to each other and to the thermal diffusivity $\lambda / \bar{c} \, \rho$
(all Lewis numbers unity), the conduction and diffusion
fluxes cancel and the convection flux is constant through
the flame. This has been discussed recently by Hirschfelder
(Ref. 16). In the data of Fig. 9, the convection term shows
a small positive maximum, but the present authors attach no
general significance to this. The sum of the three fluxes
shown as the dotted curve would ideally be constant through-
out the flame. The fact that the sum shows a maximum greater
than the convection term itself indicates that the sign of
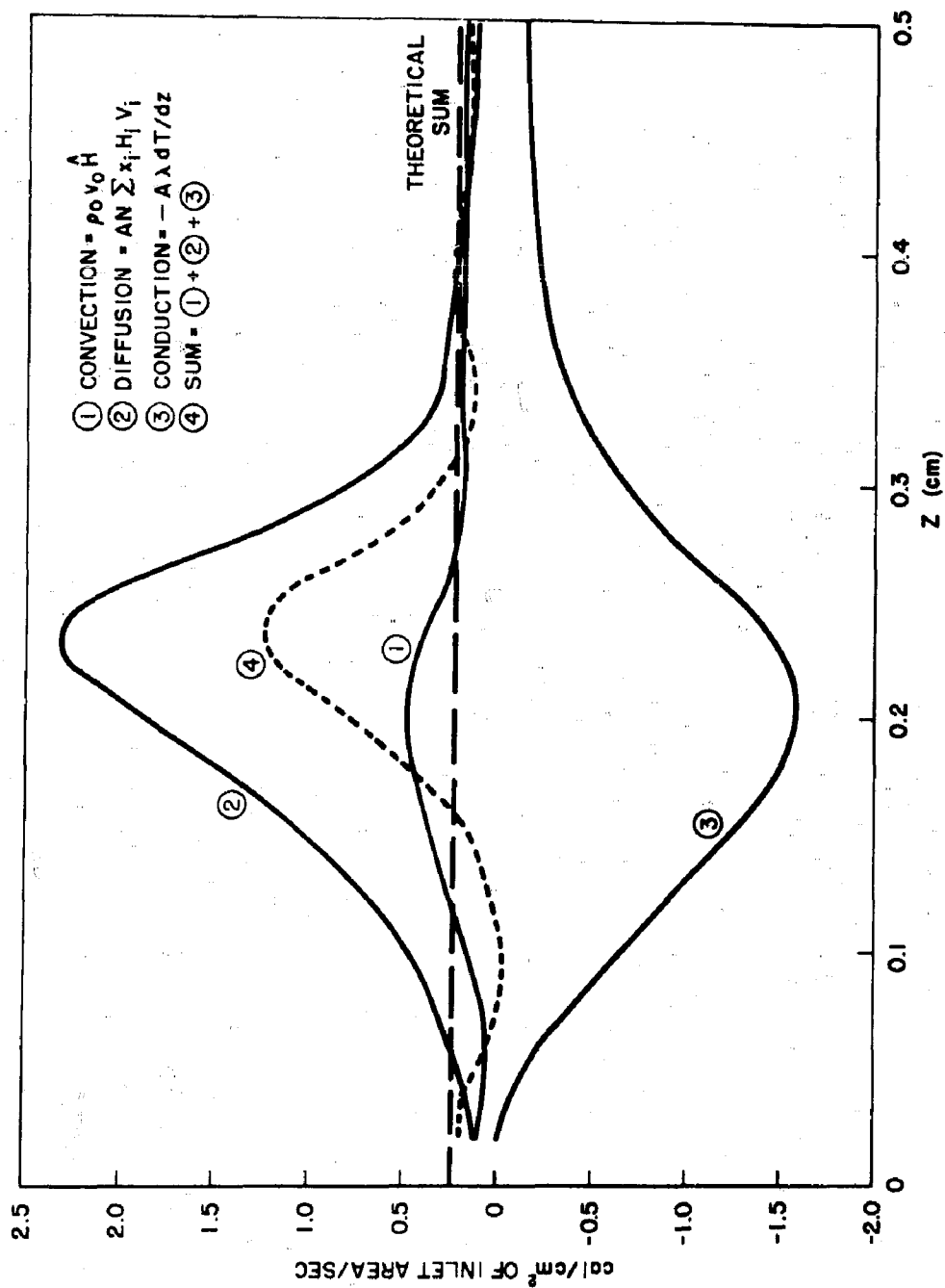the latter is not reliably established in this case.

Fig. 9   CONSERVATION OF ENERGY (ENTHALPY BALANCE) AND ENTHALPY FLUXES THROUGH FLAME ZONE

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IV.   CONCLUSION

Since no absolute criterion or prior data for
comparison exist, it is difficult to judge the energy
balance result.   It might well be that the neglect of
free radicals (which show up as stable products in
the sampled gas, of course) is more serious from an
energetic point of view than in the material balances.
At any rate, the results given in Fig. 9 seem reason-
able, and there are no gross violations of energy con-
servation. As noted in Appendix C, the actual mix-
ture thermal conductivity is probably greater than the
pure oxygen value assumed, and this would tend to make
the total energy balance somewhat better.

APPENDIX A

Binary Mixture Approximation for Diffusion

The use of rigorous, multicomponent diffusion coefficients in a mixture even as relatively simple as dealt with here is completely impractical, and (fortunately) not necessary either. For any species i present in small concentration in a mixture, it can be shown (Ref. 17) that the effective diffusion coefficient of species i is closely approximated by

$$D_{i-mix} \simeq \frac{1 - x_i M_i / \overline{M}}{\sum_{j \neq i} x_j / D_{ij}} . \tag{A1}$$

This relation has been experimentally verified (Refs. 18 and 19). If, in addition, one species k is present in such excess that all the other components may be regarded as traces, then the above simplifies still further to

$$D_{i-mix} = D_i \simeq D_{ik} , \tag{A2}$$

which is the approximation used in Eq. (4). This is certainly justified. In the flame considered here, in no case did the diffusion coefficient D_{i-O2} differ from the D_{i-mix} calculated from Eq. (A1) by more than 3 per cent, which is about the experimental error of the diffusion coefficient measurements.

APPENDIX B

Neglect of Thermal Diffusion

For any tract component, the expression for the diffusion velocity including thermal diffusion may be written (Ref. 10)

$$V_i \approx - \frac{D_i}{x_i} \left[\frac{dx_i}{dz} \pm \frac{k_T}{T} \frac{dT}{dz} \right], \tag{B1}$$

where the positive sign holds if the trace i is heavier than the main component, and the negative sign if the trace is lighter. k_T is the thermal diffusion ratio given by

$$k_V \approx \pm D_i^T / N M_i D_i, \tag{B2}$$

D_i^T being the trace thermal diffusion coefficient, and N the total molar density. In most cases, D_i^T itself is positive if the trace is heavier and negative if it is lighter than (in our case) oxygen, so that k_T is generally positive. Therefore, the common situation is for a heavier trace species to thermally diffuse upstream and a lighter species downstream.

The thermal diffusion correction to the diffusion velocity was carried out for the case of H_2O, for which it probably would be as large as for any of the major species. The necessary thermal diffusion data were computed from our concentration diffusion coefficients and viscosities as outlined by Amdur and Mason (Ref. 20). At most, the thermal diffusion contribution to V_{H_2O} was about 10 per cent. Thus, while the effect is not always as small as one would like, the error caused by its neglect is at worst probably no larger than others, and the simplification this neglect brings to an already complex analysis is most welcome.

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APPENDIX C

Pure Oxygen Approximation for Thermal Conductivity

The use of λ_{O_2} instead of the true mixture thermal conductivity was occasioned primarily (as with the other approximations) by the great simplification it afforded. The actual flame mixture was composed throughout of roughly 80 per cent oxygen, with the remaining 20 per cent mostly lighter components (except for CO_2) which might be expected to increase the thermal conductivity over that of pure oxygen. To get some idea of the error introduced here, an approximate calculation (Ref. 21) was made of λ for a mixture composed of 85 per cent oxygen and 15 per cent water at 1500°K. This gave a value about 15 per cent higher than the pure oxygen value. Such an increase in λ would improve the enthalpy balance of Fig. 9 somewhat.

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